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(9) Quarterly Letter Report <sup>no.</sup> Number 6, ✓ (1)  
(6) NEODYMIUM LASER GLASS IMPROVEMENT PROGRAM

(16) <sup>ARC</sup> Proj 548;  
ARPA Order Number 306162  
Project Code Number 7300  
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Office of Naval Research  
Navy Department  
Washington, D. C.

(11) 1964

(10) by  
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During the three month period ending 30 September 1963 the areas investigated were as follows: (1) the effects of composition of a neodymium-doped laser glass on its fluorescent lifetime, (2) an analysis of the shape of the fluorescent decay curve, (3) the quantum efficiency of the fluorescence process and (4) energy threshold measurements on laser rods made from small experimental glass melts.

This research is part of Project DEFENDER, under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research and the Department of Defense.

1. The Effect of Composition on Fluorescent Lifetime

Through analysis of extensive lifetime vs. composition data the change in lifetime with concentration ( $\Delta\tau/\Delta\text{wt.}\%$ ) for each of a number of glass ingredients, namely,  $\text{Li}_2\text{O}$ ,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ ,  $\text{Rb}_2\text{O}$ ,  $\text{CaO}$ ,  $\text{BaO}$ ,  $\text{MgO}$ ,  $\text{ZnO}$ ,  $\text{B}_2\text{O}_3$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Nd}_2\text{O}_3$  has been determined. In general, the cations appear to act independently of each other in their effect upon lifetime and attempts to predict changes in lifetime with changes in composition have met with a fair degree of success.

2. Analysis of the Shape of the Fluorescence Decay Curve

An analysis of the shape of the decay curve is being made to gain some insight into the structure of the laser glass. The approach being taken is that of resolving the observed decay curve into two or more true exponential curves which represent the fluorescent emission from Nd ions having two or more different environments. It is hoped that this type of analysis may indicate whether these environments are associated with a specific ingredient of the composition, e.g. one decay lifetime associated with a potassium-influenced Nd site and another with a barium-influenced site, etc., or whether all Nd sites in a given glass are basically identical but that statistically some Nd ions are close enough together to form a coupled site with a different lifetime.

The technique for resolving the observed lifetime into its components is the same as that used in determining the half-

lives of two or more simultaneous nuclear decay processes. At the present time we are assuming that the observed curve may be represented as the sum of just two exponentials with lifetimes  $\tau_1$  and  $\tau_2$ . At low  $\text{Nd}_2\text{O}_3$  concentrations the ratios of  $\tau_1:\tau_2$  are observed to vary from 2 to 3:1. One proposed mechanism for concentration quenching is the transfer of an excited electron from one  $\text{Nd}^{+3}$  ion to the next, in which case the sites are no longer independent of each other and the above technique is no longer valid for determining the lifetimes of different sites. As concentration quenching takes place the ratio of  $\tau_1:\tau_2$  approaches 1:1, as expected on the basis of the above postulated mechanism for concentration quenching. The value of  $\tau$  for a glass exhibiting concentration quenching is half way between the values  $\tau_1$  and  $\tau_2$  for unquenched glasses in the same series. Thus we conclude that, (1) a spacial transfer of excited electrons does take place and (2) it takes place at a rate which is fast compared to fluorescent emission transitions.

### 3. Quantum Efficiency of Fluorescence

The following approach is being taken to calculate quantum efficiency. The transition probability ( $A_{\ell,u}$ ) for absorption of the 880 line may be calculated<sup>1</sup> if the extinction is known. This was determined from absorption spectra for three types of glasses (barium crown, soda-lime and flint) with varying amounts of  $\text{Nd}_2\text{O}_3$ . The transition probability for emission at 880 m $\mu$  ( $A_{u\ell}$ ) is equal to  $A_{\ell,u}$  times the ratio of the degeneracies of the two levels  $g_\ell/g_u$ .

From this calculated value of the 880 m $\mu$  emission transition probability  $A_{(u\ell)}$  and the ratio of this probability with respect to the other transitions as obtained from the emission spectra, the total transition probability ( $A_t$ ) may be calculated. The reciprocal of  $A_t$  is  $\tau_0$ , the maximum attainable value of  $\tau$  which is obtained when all excited states are deactivated by spontaneous emission. The ratio of the actual measured value of  $\tau$  to  $\tau_0$  therefore gives a value for quantum efficiency. The initial values of quantum efficiency obtained range from about 1.5 to 1.75, i.e. the measured values are actually higher than the calculated values. Since these values are greater than one, they are obviously in error. The extinction coefficient determination will be re-examined as a possible source

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<sup>1</sup>S.J.Strickler & R.A.Berg, J. Chem. Soc. 37 No. 4, 814-822 (8-15-62)

of this error. The degeneracy ratio of the upper and lower states may also be wrong. An experimental determination of quantum efficiency is being considered to check the calculated values.

#### 4. Threshold Energy for Laser Action

The new simplified procedure devised for measuring threshold of small experimental laser glass melts is providing quite consistent results. In this procedure samples are prepared from a rod drawn to 0.080" in diameter, then scribed and broken into 6" lengths and used "as is". Gold foil caps, one with a 0.010" diameter hole in it, provide the opaque and partially transmitting end reflectors for the cavity. Using this procedure the threshold for nine different experimental 1-pound melts (2 to 4 sample rods were prepared from each melt) were measured. The threshold reproducibility for a given glass was found to vary from 0% in two cases to 10% in one case. With the old procedure threshold values for rods made from a given melt often varied by a factor of two and sometimes by an order of magnitude.

#### Requested Information:

(a) Reports published during the past 12 months:  
Lifetimes of  $\text{Nd}^{+3}$  Doped Silicate Laser Glasses  
(presented at the spring meeting of the Optical Society of America, Jacksonville, Florida, March 27, 1963). Quarterly Letter Reports Numbers 3, 4 and 5. Semi-Annual Reports Numbers 2 and 3.

(b) Roger Weiss was associated with this project for three months during the summer (1963). He completed his undergraduate work at Worcester Polytechnical Institute in the spring of 1963 and started his graduate work at the University of New Hampshire in the fall of 1963.

(c) No graduate students have earned doctorates on this contract.

(d) The principal investigator (W.P.S.) is also associated with ERDL Contract DA-44-009-ENG-5155 entitled Fiber Optics Improvement Studies.

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